

## PATENT ABSTRACTS OF JAPAN

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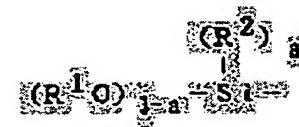
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## (54) HYDROPHILIC HARDENABLE COMPOSITION FOR BUILDING EXTERIOR AND HYDROPHILICITY IMPARTING TO BUILDING EXTERIOR SURFACE

## (57)Abstract:

PROBLEM TO BE SOLVED: To obtain the subject composition having excellent surface hardness and capable of keeping hydrophilicity even by being exposed in the outdoor for a long period of time by including a specific acrylic copolymer, etc., and making a contact angle of a hardened coating film after exposed for a specific period to water be below a specific value.

SOLUTION: This hydrophilic hardenable composition is composed of (A) 100 pts.wt. of an acrylic copolymer containing an alkoxysilyl of the formula [R<sub>1</sub> is a 1-10C alkyl; R<sub>2</sub> is H, a 1-10C alkyl or the like; (a) is 0-2], (B) 2-60 pts.wt. of a tetraalkyl silicate such as tetramethyl silicate and/or its condensate and preferably (C) a hardening catalyst (e.g. dioctyl phosphate) as an acidic hardening catalyst, and a contact angle of a hardened coating film after three months exposure with water is made to be  $\leq 52$  deg. Preferably, a using amount of the component C is 0.1-10 pts.wt. to 100 pts.wt. of solid resin of the components A and B. The component A is obtained by, e.g., copolymerizing methyl methacrylate with  $\gamma$ -methacryloxypropyltrimethoxysilane, etc.



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## CLAIMS

[Claim(s)]

[Claim 1] (A) A general formula:  
[Formula 1]



the inside of a formula, and  $\text{R}^1$  — the alkyl group of the carbon numbers 1~10, and  $\text{R}^2$  — a hydrogen atom or the alkyl group of the carbon numbers 1~10. The univalent hydrocarbon group chosen from the group which consists of an aryl group and an aralkyl group, a — 0, 1, or 2 — being shown — the alkoxy-silyl-groups content acrylic copolymer containing the basis expressed — 100 weight section. (B) The construction exterior Mochichika aquosity hardenability constituent which consists of tetraalkyl silicate and/or its two to condensate 60 weight section, and a (C) curing catalyst, and is characterized by an angle of contact with the water after three-month exposure of a cured film being 52 degrees or less.

[Claim 2] The construction exterior Mochichika aquosity hardenability constituent according to claim 1 whose rate of an alkoxy-silyl-groups content monomer in the aforementioned (A) ingredient it is the polymer in which the aforementioned (A) ingredient used as a copolymerization ingredient an alkoxy-silyl-groups content monomer which contains a polymerization nature double bond in intramolecular, and is 5 to 90 % of the weight.

[Claim 3] The construction exterior Mochichika aquosity hardenability constituent according to claim 1 or 2 whose aforementioned (C) ingredient is an acid curing catalyst.

[Claim 4] The construction exterior Mochichika aquosity hardenability constituent according to claim 3 in which said acid curing catalyst is an acid anhydride of a mixture of alkyl acid phosphate, alkyl acid phosphate, and amine or a reactant, saturation or unsaturation polyvalent carboxylic acid, saturation, or unsaturation polyvalent carboxylic acid.

[Claim 5] A construction exterior Mochichika aquosity hardenability constituent given in any 1 paragraph of claims 1~4 by which methacrylic acid n butyl is contained in the aforementioned (A) ingredient as a copolymerization monomer.

[Claim 6] An angle of contact with water after three-month exposure forms a cured film of 52 degrees or less in the construction exterior surface, give hydrophilic nature to the construction exterior surface concerned, and by this A giving hydrophilic characteristic method for the construction exterior surface which is a method which make a pollutant adhering to the construction exterior surface easy to flush with storm sewage etc., and is characterized by applying a construction exterior Mochichika aquosity hardenability constituent of claim 1~5 given in any 1 paragraph to the surface of the construction exterior.

[Translation done]

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## DETAILED DESCRIPTION

## [Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to a construction exterior Mochichika aquosity hardenability constituent (hardenability constituent for top coat used for the construction exterior), and the giving hydrophilic characteristic method for the construction exterior surface.

[0002]

[Description of the Prior Art] Conventionally, the constituent containing acrylic silicon resin was covered on the surface of industrial products, such as a ceramic industry system raw material, steel, construction, and building materials. The high hardness film was also able to form that the design effect can be given to said industrial product in the surface with last thing, and, thereby, it was able to raise physical properties, such as the weatherability of said industrial product, and corrosion resistance.

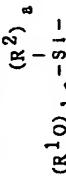
[0003] If it is furthermore in today, it is socially required for the following reasons that hydrophilic nature should be given to a membrane surface. That is, for example, contamination of the building has been a problem centering on the city part. However, by improving surface wettability, the pollutant adhering to the surface can be washed with storm sewage etc., and the resistance to contamination of said building can be raised.

[0004] Although it could realize by the method of adding a surface-active agent to said constituent, in a giving hydrophilic characteristic, the fall of surface hardness was caused, or there was a problem in the hydrophilic maintenance by the outdoor exposure over a long period of time, and it was not a still satisfying method.

[0005] This invention is made in view of the above-mentioned actual condition, and the purpose, In order that the pollutant which is in the place which provides the hydrophilic hardenability constituent which can hold hydrophilic nature, and adhered to the construction exterior surface may carry out that it is easy to washed away with storm sewage etc. even if it carries out an outdoor exposure over a long period of time while having the outstanding surface hardness, it is in the place which provides the method of giving hydrophilic nature to the construction exterior surface concerned using said hydrophilic hardenability constituent.

[0006] [Means for Solving the Problem] A construction exterior Mochichika aquosity hardenability constituent of this invention is the (A) general formula;

[Formula 2]



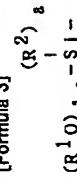
the inside of a formula, and R<sup>1</sup> — the alkyl group of the carbon numbers 1-10, and R<sup>2</sup> — a hydrogen atom or the alkyl group of the carbon numbers 1-10. The univalent hydrocarbon group chosen from the group which consists of an aryl group and an aralkyl group, a — 0, 1, or 2 — being shown — the basis expressed is contained — alkoxy-silyl-groups content acrylic copolymer (00 weight section). It consists of (it is only hereafter called a "part"), (B) tetraalkyl silicate and/or its condensate 2 — 60 copies and the (C) curing catalyst, and is characterized by an angle of contact with the water after three-month exposure of a cured film being 52 degrees or less.

[0007] A giving hydrophilic characteristic method for the construction exterior surface of this invention, 2.\*\*\*\*\* shows the word which can not be translated.

An angle of contact with water after three-month exposure forms a cured film of 52 degrees or less in the construction exterior surface, give hydrophilic nature to the construction exterior surface concerned, and by this, It is a method which make a pollutant adhering to the construction exterior surface easy to flush with storm sewage etc. and is the method of applying the above-mentioned construction exterior Mochichika aquosity hardenability constituent to the surface of the construction exterior.

[0008] [Embodying of the Invention] the alkoxy-silyl-groups content acrylic copolymer (the following and "an alkoxy-silyl-groups content acrylic copolymer (A)" — or it is also only called "(A) ingredient") of the (A) ingredient which is one ingredient in the constituent of this invention — general formula;

[Formula 3]



It is a polymer which comes out and has preferably two or more alkoxy silyl groups expressed in [ at least one ] one molecule. This alkoxy silyl groups may be contained at the end of the main chain of the (A) ingredient, may be contained in a side chain, and may be contained to both sides.

[0009] (A) The solvent resistance of a hardened material (for example, coat) in which the number of alkoxy silyl groups in one molecule of ingredients is obtained from a constituent of this invention by less than one piece falls easily.

[0010] (B) the inside of said formula, and R<sup>1</sup> — the carbon numbers 1-10 — it is an alkyl group of 1-4 preferably. If a carbon number exceeds 10, the reactivity of alkoxy silyl groups falls, and also when R<sup>1</sup> is a phenyl group and benzyl, for example except an alkyl group, reactivity will fall.

[0011] (C) An example of R<sup>1</sup>, a methyl group, an ethyl group, n-propyl group, an isopropyl group, n-butyl group, an isobutyl group, etc. are mentioned, for example,

[0012] R<sup>2</sup> is a hydrogen atom or the carbon numbers 1-10, and the univalent hydrocarbon group preferably chosen from a group which consists of an alkyl group of 1-4, an aryl group, and an aralkyl group among said formula.

[0013] The same basis as R<sup>1</sup> is raised as an example of an alkyl group which is R<sup>2</sup>, a phenyl group etc. are mentioned as an example of an aryl group, for example, and benzyl etc. are mentioned as an example of an aralkyl group, for example.

[0014] (A) basis contained in an alkoxy-silyl-groups content monomer mentioned later, for example as an example of alkoxy silyl groups expressed with said general formula [the "izing 3] is mentioned.

[0015] Since the main chain consists of an acrylic copolymerization chain substantially, an alkoxy-silyl-groups content acrylic copolymer (A) is excellent in the weatherability of a hardened material, chemical resistance, a water resisting property, etc. In the (A) ingredient, if alkoxy silyl groups has combined with a carbon atom, the water resisting property of a hardened material obtained will become the further outstanding thing, and will become what was excellent in alkali resistance, acid resistance, etc.

[0016] As for a number average molecular weight of an alkoxy-silyl-groups content acrylic copolymer (A), 1,000-30,000 are preferred from a point of physical properties, such as the endurance etc. of a hardened material obtained from a constituent of this invention, and 3,000-25,000 are still more preferred.

[0017] (A) An ingredient can be obtained, for example by copolymerization of acrylic system monomers, such as acrylic acid, methacrylic acid, and those derivatives, and an alkoxy-silyl-groups content monomer.

[0018] There is no limitation in particular in an acrylic system monomer, and as the example, Methyl (meta) acrylate, ethyl (meta) acrylate, butyl (meta) acrylate, 2-ethylhexyl (meta) acrylate, stearyl (meta) acrylate, Benzyl (meta) acrylate, cyclohexyl (meta) acrylate, Trifluoroethyl (meta) acrylate, pentfluoro propyl (meta) acrylate, Perfluoro cyclohexyl (meta) acrylate, acrylonitrile (meta) acrylate, Diethylaminoethyl (meta) acrylate, Dimethylaminoethyl (meta) acrylate, Diethylaminoethyl (meta) acrylate, acrylamide, N,N-dimethylacrylamide, N- $\alpha$ -alpha-ethyl(methyl) acrylamide, N-butoxymethyl(methyl) acrylamide, acryloyl morpholine, 2-hydroxypropyl (meta) acrylate, 2-hydroxyethoxy (meta) acrylate, and Toagosei Chemical AS-6 which are ARONIKUSU M-5700 made from N-methyl(methyl)acrylamide and Toagosei Chemical industry, and a macro monomer made from Toagosei Chemical industry, AN-6, AA-6, AB-6, Placed by

AK-5 and Daicel Chemical Industries, Ltd. FA-1, Placel FA-4, and Placel. The hydroxy alkyl ester of alpha, such as FM-1, Placel FM-4, and hydroxy alkyl ester of acrylic acid (meta), and beta-ethylenic unsaturated carboxylic acid, phosphoric acid, or a condensation product slack phosphoester group content vinyl system compound of phosphoric ester. Acrylate including a urethane bond and a siloxane bond (meta) are mentioned.

[0019] there is no limitation in particular except [ of having a polymerization nature double bond as said alkoxy-silyl-groups content monomer ] — as the example — for example [Formula 4]



usually preferred to 100 copies of resin solid content of the (A) ingredient and the (B) ingredient, and 0.1–10 copies are still more preferred. (C) When there is a tendency for hardenability to fall if the amount of ingredient used will be less than 0.1 copy and it exceeds 20 copies, there is a tendency for appearance-of-film nature to fall.

[0035] Even if it carries out repeated use over a long period of time, in order to secure satisfactory preservation stability, it is preferred to use a dehydrator and alkyl alcohol. [0036] As an example of a dehydrator, for example Methyl orthoformate, ethyl orthoformate, ester compounds, such as altimethyl acetate, alethyl acetate, methyl trimetoxysilane, gamma-methacryloxypropyl trimetoxysilane, vinyltrimetoxysilane, methylsilicate, and ethyl silicate, are mentioned. Low-molecular-weight alcohol like methanol and ethanol as alkyl alcohol is mentioned. A dehydrator and alkyl alcohol may be added before a polymerization of an alkoxy-silyl-groups content polymer (A), may be added after a polymerization, and may be added during a polymerization.

[0037] Although there is no limitation in particular in the amount of a dehydrator and the alkyl alcohol used, 0.5–20 copies two – ten copies are still more preferred to 100 copies of resin solid content of the (A) ingredient and the (B) ingredient.

[0038] If a dehydrator and alkyl alcohol are used together, an effect remarkable in preservation stability will be seen.

[0039] In a constituent of this invention, according to a use, a diluent, paints (an extender is included), Additive agents, such as an ultraviolet ray absorber, light stabilizer, an antisetting agent, and a leveling agent; A nitrocellulose, Fibrin, such as cellulose acetate butyrate; resin; bulking agents, such as an epoxy resin, melamine resin, vinyl chloride resin, chlorinated polypropylene, chlorinated rubber, and a polyvinyl butyral, etc. may be added.

[0040] After making the above-mentioned constituent a coated object with \*\* with conventional methods, such as various paint especially immersion, spraying, and brush coating, a coat which was excellent in adhesion, endurance, etc. on the surface of a coated object can be formed by making it usually harden above 30 \*\*.

[0041] [Example] Hereafter, the construction exterior Mochichika aquosity hardenability constituent of this invention is explained still more concretely based on an example.

[0042] After carrying out temperature up to 110 \*\*, having taught 40.4 copies of xylene to the reaction vessel provided with synthetic example 1 agitator, the thermometer, the reflux condenser, the nitrogen gas introducing pipe, and the dropping funnel, and introducing nitrogen gas into it, uniform dropping of the mixture (a) of the following presentation was carried out over 5 hours with the dropping funnel.

mixture (a)  
Methyl methacrylate 28.4 copies Methacrylic acid n BUCHI 47.1 copies Butyl acrylate 11.8 copies Gamma-methacryloxypropyl trimethoxy silane 11.8 copies Acrylamide 0.9 copy Xylene 17.9 copies 2-azobisisobutyronitrile 1.0 copy.

[0043] Uniform dropping of 0.5 copy of 2,2-azobisisobutyronitrile and 8.1 copies of toluene was carried out over 1 hour after the end of dropping of a mixture (a). It cooled after 2-hour aging at 110 \*\* after the end of dropping, xylene was added to the resin solution, and solids concentration was adjusted to 50%. The number average molecular weight of the obtained resin was 15,000.

[0044] After carrying out temperature up to 110 \*\*, having taught 40.4 copies of xylene to the reaction vessel provided with synthetic example 2 agitator, the thermometer, the reflux condenser, the nitrogen gas introducing pipe, and the dropping funnel, and introducing nitrogen gas into it, uniform dropping of the mixture (b) of the following presentation was carried out over 5 hours with the dropping funnel.

mixture (b)  
Methyl methacrylate 56.6 copies Butyl acrylate 30.7 copies Gamma-methacryloxypropyl trimethoxy silane 11.8 copies Acrylamide 0.9 copy Xylene 17.9 copies 2,2-azobisisobutyronitrile 1.0 copy.

[0045] Uniform dropping of 0.5 copy of 2,2-azobisisobutyronitrile and 8.1 copies of toluene was carried out over 1 hour after the end of dropping of a mixture (b). It cooled after 2-hour aging at 110 \*\* after the end of dropping, xylene was added to the resin solution, and solids concentration was adjusted to 50%. The number average molecular weight of the obtained resin was 15,000.

[0046] After carrying out temperature up to 110 \*\*, having taught 30 copies and 10.4 copies of xylene to the reaction vessel provided with synthetic example 3 agitator, the thermometer, the reflux condenser, the nitrogen gas introducing pipe, and the dropping funnel for ES140 (\*), and introducing nitrogen gas into it, uniform dropping of the mixture (c) of the following presentation was carried out over 5 hours with it, uniform dropping of the mixture (c) of the following presentation was carried out over 5 hours with



Epoxy compounds, such as car DEYURAE made from Oil recovery Shell Epoxy, Epicoat 828 made from Oil recovery Shell Epoxy, and Epicoat 1001, phosphoric acid, and/or an addition reaction thing with monoalkyl acid phosphate, Acidic compounds, such as maleic acid, adipic acid, azelaic acid, sebacic acid, itaconic acid, citrate, succinic acid, phthalic acid, trimellitic acid, pyromellitic acid, these acid anhydrides, Para toluenesulfonic acid, and dodecylbenzenesulfonic acid, are mentioned. The mixture or reactant of these acid catalysts and amine is also contained. For example, amines, such as hexylamine, N,N-dimethylidodecyl amine, and dodecyl amine, are mentioned.

[0033] An acid curing catalyst is desirable among these curing catalysts (C), mixture or reactant of activity of alkyl acid phosphate, alkyl acid phosphate, and amine is high, and hydrophilic nature of a reactant is also especially preferred. A curing catalyst (C) may be used independently and may use two or more sorts together.

[0034] (C) Although there is no limitation in particular in the amount of ingredient used, 0.1–20 copies are

